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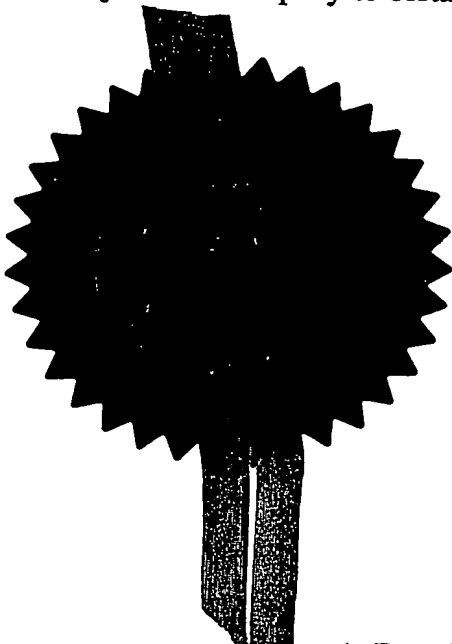
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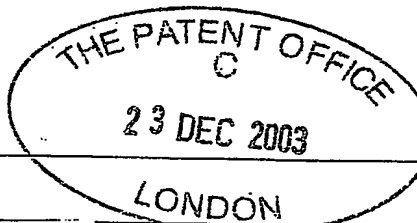
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*Stephen Hordley*

Dated 5 May 2004

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1. Your reference

JPP228

2. Patent application number  
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0329836.1

23 DEC 2003

3. Full name, address and postcode of the or of  
each applicant (underline all surnames)BAC2 Ltd  
27 Stubbington Way  
Fair Oak  
Eastleigh  
Hants  
SO50 7LQ24DEC03 E361814-1 D02806  
F01/7700 0.00-0329836.1 NONE

Patents ADP number (if you know it)

8353344001

If the applicant is a corporate body, give the  
country/state of its incorporation

United Kingdom (GB)

4. Title of the invention

SOLID POLYMER ELECTRODE

5. Name of your agent (if you have one)

Barker Brettell

"Address for service" in the United Kingdom  
to which all correspondence should be sent  
(including the postcode)10-12 Priests Bridge  
London  
SW15 5JE

7442494003

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6. Priority: Complete this section if you are  
declaring priority from one or more earlier  
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Country

United  
Kingdom  
(GB)Priority application number  
(if you know it)

0308135.3

Date of Filing  
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9 April 2003

7. Divisionals, etc: Complete this section only if  
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## SOLID POLYMER ELECTRODE

The present invention relates to a solid polymer electrode, particularly a carbon electrode.

5

Carbon electrode materials are used in many electrochemical applications and industrial processes including fuel cells, batteries, energy storage capacitors, catalyst support, corrosion control of metals and concrete, metal production and water treatment (e.g. water purification, 10 desalination, metal recovery from industrial waste, de-ionisation, electrochemical destruction of pollutants and contaminants from water in water treatments such as water softening and pH control).

Electrochemical water treatment processes are becoming increasingly of 15 interest as they offer benefits over conventional physical, chemical and biological systems. Being electrically driven, rapid reaction rates can be achieved enabling smaller and more efficient plant design. Electrochemical methods replace the need for chemical treatment and biological systems and have been shown to produce other benefits such as 20 a reduction in sludge production and a reduced need to transport hazardous chemicals.

A 2002 report from Water UK states (ENDS report issue 339 1/4/03) that water treatment in the UK is becoming more expensive due to the 25 increased quality standards. There is an increase in energy use for both drinking and wastewater treatment. Drinking water now requires 600 kilowatt hours per megalitre to treat and supply - up by 28% since 1998 due to additional treatment and pumping required to meet the cryptosporidium regulations. On the wastewater side, energy use has also 30 escalated from 437kWh/Ml in 1998 to 598kWh/Ml - an increase of 37%. The reasons for this are a host of increasing environmental quality

requirements obliging companies to install more secondary treatment and ultraviolet disinfection, and processing and disposal of increasing quantities of sewage sludge.

- 5 The need for more efficient, lower cost water treatment is clear. Although not widespread, electrochemical water treatment systems have been developed and commercialised. Existing carbon materials, particularly 3-dimensional carbon electrodes, used in electrodes are expensive to produce as high temperature carbonising steps are involved.
- 10 The resulting products are typically brittle, fragile and require machining to the design shape.

The basic operating principles of carbon electrodes are readily understood, but the manufacturing techniques for producing carbon electrode material are limited. US patent 6544648 gives a description of a high-pressure process to manufacture porous carbon electrodes. The following paragraphs summarise the other known production processes used to produce carbon materials for electrode use.

- 20 Three processes are currently used, identified by the types of materials they employ as feedstock: granular activated carbon, carbonisation of polymers, and carbon aerogels. Traditionally carbon electrodes are produced with an organic or polymeric binder mixed together with the carbon powders. A disadvantage exists with the use of binders to form the electrodes. As most binders are non-conductive then the conductivity of the electrode deteriorates.

Modern carbon electrodes are manufactured from thermosetting resins by a process in which the resin is pre-formed to a certain shape then subjected to high temperatures for extended periods of time until complete carbonisation occurs. The volume of carbon formed is considerably

smaller than the original resin size, which leads to reduced product yield. This is a significant problem if specific geometric shapes or sizes are required. This manufacturing technique also has the disadvantages of high material cost and weak material strength due to the "shrinking" of the precursor carbon at high carbonisation temperatures.

Some specific carbon electrodes are manufactured from aerogel compounds with sol-gel technology by carbonising organic compounds. The pyrolysis process produces a vitreous carbon material, which has a high surface area and high electrical conductivity. However, this manufacturing technique includes extremely high manufacturing costs. The final shape of the carbon materials is much smaller than the feed material. Additional processing would be required to produce a specific geometric shape.

The production techniques used to manufacture carbon electrodes can be improved and there exists a need for a more efficient, less expensive and more flexible process to manufacture carbon electrodes.

A solution to these problems has been sought.

According to the invention there is provided a separator plate suitable for use in a fuel cell having one or more flow field for directing gas flow wherein the plate comprises a carbon-doped ester-cured alkaline phenolic resole resin containing conducting alkaline salts.

According to the invention there is also provided an electrical device comprising:

- (a) a negative electrode;
- (b) a positive electrode; and
- (c) an electrolyte means; and optionally

(d) a separator and/or bipolar plate;

wherein one or more electrode and/or separator or bipolar plate comprises a carbon-doped ester-cured alkaline phenolic resole resin containing conducting alkaline salts.

5

The bipolar and/or separator plate used in the electrical device according to the invention is preferably a separator plate according to the invention.

According to the invention there is further provided an electrode  
10 comprising a carbon-doped ester-cured alkaline phenolic resole resin containing conducting alkaline salts.

Cured phenolic resins are thermoset polymers and are superior to all  
other resin systems with respect to their good thermal and mechanical  
15 stability, and their flame resistance. Normally they have good electrical insulating capabilities too. It is therefore surprising that an ester-cured alkaline resole resin is useful as an electrode.

The advantages of using a carbon-doped ester-cured alkaline phenolic  
20 resole resin containing conducting alkaline salts to form one or more of the electrodes in the cell according to the invention include that

- the resin can be moulded at room temperature such that the production process is simpler and less costly;
- the resin is of higher conductivity than binders normally used to  
25 bind carbon so deterioration in conductivity is minimised;
- the resin is room temperature cured with minimal shrinkage resulting in a more durable product requiring no machining and having good material strength;
- the resin is infinitely water dilutable and as such added water can  
30 improve wetting of carbon or graphite powders or flakes to enable high carbon content electrode material to be produced;

- the resin can be foamed to produce a 3-d porous carbon structure with high surface area;
- the resin can be doped with conductive fillers: and
- the starting materials are relatively cheap producing cost savings.

5

To illustrate the cost saving, the following figures are provided. A commercially available carbon RF [resorcinol-formaldehyde] aerogel material is supplied in the form of a block, granules, powders and papers. In November 2003, a 100g quantity of RF aerogel costs US\$185 before  
10 machining. RF aerogel paper sheets (3.5 inches x 10 x 0.01) cost about US\$665 per 100 sheets. Carbon aerogels are more expensive with 100g costing \$275 and 100 paper sheets costing \$900. In comparison, 100g of a moulded form of the carbon doped resin material used in the invention would cost around US\$0.42.

15

For a lower cost electrode material such as graphite and copper the material cost represents only a small part of the total electrode production costs. Graphite and copper require high temperature machining tools to produce precision shapes. Production time, machining time, labour and  
20 scrap are all critical to the overall costs. The carbon doped resin used in the present invention is advantageous because it can be moulded and hardened to a precise shape without machining, reducing labour and scrap.

25 As a result of the costs savings provided with the cells according to the invention, it may be economic to use industrial electrochemical processes that, at present, are not cost competitive with other water purification techniques such as chlorination, ozonation and coagulation.

30 The electrical device according to the invention is preferably a cell, a battery including two or more cells, or a capacitor (especially an



electrolytic capacitor). Where the electrical device is a fuel cell, it includes at least one bipolar plate and inlets and outlets to control flow of oxygen and hydrogen through the cell.

- 5 A separator according to the invention has a single flow field. It is useful as a current collector, particularly in a fuel cell. A bipolar plate is a flat, gas impermeable, electrically conductive separator between individual fuel cells in a stack. It has a flow field on each side. A flow field is preferably at least one channel machined or molded into the plate. The  
10 flow field is suitable for carrying fuel (usually hydrogen) on one side and an oxidant on the other side from entry and exit points in the fuel cell.

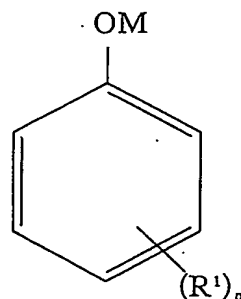
The electrolyte means of the electrical device according to the invention is optionally either in the form of an electrolyte or it is arranged to  
15 receive an electrolyte. For example the electrolyte means could be in the form of a conduit through which an electrolyte could flow during operation of the cell or the electrolyte means could be in the form of a container into which an electrolyte could be placed at least during operation of the cell.

20

The invention also provides use of a carbon-doped ester-cured salt-containing alkaline resole resin as an electrode.

The carbon-doped ester-cured salt-containing alkaline phenolic resole  
25 resin is preferably a reaction product of an ester curing agent with a phenolic resole and a base. The phenolic resole is preferably a reaction product of a phenol-reactive aldehyde with an alkaline compound of formula

30



(I)

5

wherein R<sup>1</sup> is a straight or branched chain optionally unsaturated alkyl group containing from 1 to 8 carbon atoms (preferably from 1 to 4 carbon atoms, more preferably from 1 to 2 carbon atoms) optionally substituted by a halogen atom (preferably chlorine) or a hydroxy group, a halogen atom (preferably chlorine), a hydroxy group, and/or a phenyl or benzyl group (optionally substituted by a hydroxy group and/or a straight or branched chain alkyl group containing from 1 to 8 carbon atoms (preferably from 1 to 4 carbon atoms, more preferably from 1 to 2 carbon atoms) optionally substituted by a halogen atom (preferably chlorine) or a hydroxy group);

M is a mixture of hydrogen ions and at least one further cation (preferably the at least one further cation is an alkali metal cation (preferably sodium, lithium or potassium), an alkaline earth metal cation (preferably barium, magnesium or calcium), and/or a N(R<sub>2</sub>)<sub>4</sub><sup>+</sup> ion. (wherein each R<sub>2</sub> is the same or different and is hydrogen or a straight or branched chain alkyl group containing from 1 to 4 carbon atoms)) wherein the molar ratio of hydrogen ions to the at least one further cation is sufficient for the pH to be greater than 7 and is preferably from 2:1 to 1:1; and  
n is from 0 to 2.

Examples of suitable compounds of formula (I) include, but are not restricted to the salt of phenol itself, salts of substituted phenols such as alkylated phenols, halogenated phenols and polyhydric phenols, and

hydroxy-substituted poly-nuclear aromatics. Examples of alkylated phenols include methylphenol (also known as cresol), dimethylphenol (also known as xlenol), 2-ethylphenol, pentylphenol and tert-butyl phenol. Examples of halogenated phenols are chlorophenol and bromophenol. Examples of polyhydric phenols include 1,3-benzenediol (also known as resorcinol), 1,2-benzenediol (also known as pyrocatechol), 1,4-benzenediol (also known as hydroquinone), 1,2,3-benzenetriol (also known as pyrogallol), 1,3,5-benzenetriol and 4-tert-butyl-1,2-benzenediol (also known as tert-butyl catechol). Examples of hydroxy-substituted poly-nuclear aromatics include 4,4'-isopropylidenebisphenol (also known as bisphenol A), 4,4'-methylidenebisphenol (also known as bisphenol F) and naphthol.

Salts of compounds formed by the condensation reaction of two or more compounds of formula (I) with one or more molecules of a phenol-reactive aldehyde are suitable for use in the ester-cured alkaline resole resin. Examples include, but are not limited to, resinous reaction products of phenol itself, salts of substituted phenols such as alkylated phenols, halogenated phenols and multi-hydroxy phenols, and hydroxy-substituted multi-ring aromatics. Furthermore, mixtures of aldehyde-reactive phenols, such as those obtained from coal tar fractionation, depolymerised lignin and cashew nut shell liquid, can be employed as all or part of the resole component.

The phenol-reactive aldehyde used to react with the compound of formula (I) to form an alkaline phenolic resole is preferably a compound of formula



(II)

wherein R represents a hydrogen atom or a straight or branched chain alkyl group having from 1 to 8 (preferably from 1 to 4, more preferably from 1 to 2, most preferably 1) carbon atoms; or a precursor of a compound of formula (II).

5

Examples of suitable aldehydes include formaldehyde, acetaldehyde, propionaldehyde, n-butylaldehyde, n-valeraldehyde, caproaldehyde. Compounds suitable for use as precursors for a compound of formula (II) include compounds that decompose to formaldehyde such as  
10 paraformaldehyde, trioxane, furfural, hexamethylenetriamine, acetals that liberate formaldehyde on heating, and benzaldehyde.

The aldehyde is preferably reacted with the compound of formula (I) in a ratio of from 1:1 to 1:3, preferably from 1:1.2 to 1:3, more preferably  
15 from 1:1.5 to 1:3.

Some of the alkalis  $M(OH)_x$  (where M represents a non-hydrogen cation and x represents 1 or 2) are not very soluble in an aqueous resin e.g. calcium hydroxide. They can still be used by dehydrating the resin and  
20 using the ester as a solvent for the resole. The water insoluble alkali can then be dispersed in the resin to form a paste. A polar solvent (for example water) is then required to start the reaction.

The ester curing agent used to cure the alkaline phenolic resole resin is  
25 preferably of formula



wherein  $R^3$  represents a hydrogen atom or a straight or branched chain  
30 alkyl group containing from 1 to 8 carbon atoms (preferably from 1 to 4,

more preferably from 1 to 2 carbon atoms) optionally substituted by a halogen atom; and

R<sup>4</sup> represents a straight or branched chain alkyl group containing from 1 to 8 carbon atoms (preferably from 1 to 4, more preferably from 1 to 2 carbon atoms) optionally substituted by one or more hydroxy and/or R<sup>3</sup>COO- groups, or

a phenyl group optionally substituted by a straight or branched chain optionally unsaturated alkyl group containing from 1 to 8 carbon atoms (preferably from 1 to 4 carbon atoms, more preferably from 1 to 2 carbon atoms) optionally substituted by a hydroxy group, a halogen atom (preferably chlorine), a hydroxy group, and/or a phenyl or benzyl group (optionally substituted by a hydroxy group and/or a straight or branched chain alkyl group containing from 1 to 8 carbon atoms (preferably from 1 to 4 carbon atoms, more preferably from 1 to 2 carbon atoms)); or

R<sup>3</sup> represents a chemical bond to R<sup>4</sup> and R<sup>4</sup> represents a straight or branched chain alkyl group containing from 2 to 10 carbon atoms (preferably from 2 to 4 carbon atoms).

The ester curing agent for the alkaline resole resin must be reactive with the alkali to produce a salt in the cured resin. Reactive esters can include, but are not restricted to, carboxylic acid esters, esters of polyhydric alcohols, lactones and carbonate esters, phenolic esters and resole esters. Examples of reactive carboxylic acid esters are methyl formate and ethyl formate. Examples of reactive polyhydric alcohol esters which may be used as curing agent for the resin include glycerol triacetate and ethylene glycol diacetate. Examples of reactive carbonate esters include cyclic carbonate esters such as propylene carbonate and ethylene carbonate. Examples of reactive lactones include propiolactone, butyrolactone, valerolactone and caprolactone. Examples of reactive phenolic esters are phenyl acetate and resorcinol diacetate. An example of a reactive resole ester is 2,4,6-tris-acetoxymethylphenyl acetate.

Mixtures of esters may be used for example propylene carbonate and triacetin to vary the rate of cure.

5 It has been found that the rate of curing of the resole by the ester is determined primarily by the acidity of the conjugate acid; e.g. ethyl formate ( $R^3 = H$ ) reacts approximately 1000 times faster than ethyl acetate ( $R^3 = CH_3$ ) due to the greater acidity of formic acid over acetic acid. The gel times achieved using each ester also exhibit a similar order of difference. The carbon chain length of the alcohol ( $R^4$ ) influences saponification rates and gel times to a lesser extent with the saponification rate reduced and the gel time increased with each additional carbon. It is also been found that as the chain length and/or branching of  $R^3$  and  $R^4$  increases the miscibility of the resin and ester is reduced. Good compatibility between the ester and resin is essential for the cure reaction to proceed. It is therefore clear that the selection of an ester curing agent will determine the cure rate of the reaction, and will also determine the carboxylate ion formed in the reaction and ultimately the salt contained in the polymer matrix which influence the properties of the electrode.

20 The base used in the reaction to form the ester-cured salt-containing alkaline phenolic resole resin used in the invention is preferably an alkaline compound which is capable of forming a conducting salt which is soluble in the resin used in the invention. An example of a suitable base is a hydroxide or an oxide of an alkali or alkaline earth metal or of ammonia, e.g. lithium, sodium, potassium, magnesium, calcium, barium or ammonia.

The ester-cured salt-containing alkaline phenolic resole resin used in the invention is prepared from a resole resin, an ester curing agent, one or more bases and, optionally, a polar solvent. Methods for their synthesis are well known to a person of skill in the art and are described in DE-C-1

065 605, DE-C-1 171 606, JP-A 49-16793 and JP-A 50-130627. According to these publications, a highly alkaline phenolic resole resin in aqueous solution may be cured at ambient temperature by reaction with an organic ester by contacting the resin with the ester in the form of a liquid or a gas. The ester-cured alkaline phenolic resole resin containing conducting alkaline salts used in the invention is optionally in dried or in anhydrous form depending on the particular properties required.

Ester cured alkaline phenolic resoles can be differentiated from acid cured resoles in that the polymer matrix of the cured alkaline phenolic composition contains a high level of alkaline salts. To disperse or dissolve salts of carboxylic acids is very difficult in liquid resoles as the phenolic resin can lose solubility and precipitate from solution. Secondly, acidifying a phenolic resole containing dispersed salts of carboxylic acids will generate  $\text{CO}_2$  due to the decomposition of the salt on reaction with the acid. It is therefore a unique feature of the cured alkaline resole composition that high quantities of carboxylic acid salts are present in the cured polymer matrix having been formed during the cure reaction.

The form of carbon selected to improve conductivity is preferably graphite powder or flake. The main requirements of the carbon is resin compatibility, carbon wettability and conductivity. More than one carbon combination can be used with blends of different carbons such as activated carbon powder and other conductive carbon powders. Carbon combinations with non-carbon conductive fillers such as metal and metal oxide powders and metal coated graphite and glass such as nickel coated graphite and silver coated glass.

An additional feature of ester cured alkaline phenolic resins is that unlike most common thermosetting polymer systems they are aqueous based and

infinitely dilutable with water. The polymerisation reaction on addition of the ester curing agent to a water diluted resin is unaffected and hardening of the polymer proceeds although at a slower rate. Use of a faster curing ester will however speed up the hardening process to a desirable rate. The main advantage water dilutability imparts to a carbon doped ester cured phenolic resin mixture is the ability to wet the carbon or graphite powders or flakes to enable high carbon content electrode material to be produced and consequently higher conducting electrodes formed from the material. On room temperature curing the added water evaporates from the formed electrode material and a hard, solid, high carbon, low water, carbon doped ester cured phenolic polymer results.

A controlled addition of water to the resin/carbon mixture is an important processing feature as the consistency of the mixture can be changed to suit the room temperature injection moulding, pouring or casting techniques employed. At high water additions control of the mixture consistency can be enhanced by the inclusion of a thickener such as a starch or a starch derivative, cellulose or a cellulose derivative, a natural gum such as gum arabic or guar gum or a synthetic thickening agent such as a polyamide or a polyacrylate.

The resin used in the invention optionally includes a plasticiser to increase flexibility of the resin. It will be appreciated that for some applications a flexible resin would be useful. The plasticiser is preferably inert, alkali compatible, non-volatile, and/or liquid. Preferably the plasticiser is soluble in the resin and/or the ester curing agent. Levels of plasticiser are determined by the application requirements and are limited by effects on conductivity of the cured electrode material. Examples of the plasticiser include an excess of the ester curing agent, polyvinylacetate and/or a polyethylene glycol.



Ester cured phenolic resins may be foamed by employing a foam blowing agent in the resin formulation to form a 3-d porous structure. Examples of foam blowing agents can include any low boiling solvent of low water miscibility such as trichloromonofluoromethane (CFC-11), hydrogenated  
5 chlorofluorocarbons (called "HCFCs"), partially hydrogenated fluorocarbons (called "HFCs"), hydrocarbons such as iso-pentane and cyclopentane. The use of blowing agents in this application is not to produce insulating foam but only to form a 3-d open cell structure therefore blowing agents are lost during the curing stage and can be  
10 recovered and recycled after formation of the 3-d structure. Other blowing such as carbon dioxide and nitrogen can be employed.

The invention is illustrated by reference to the following Figures of the drawings in which Figures 1 and 2 show results from tests using a cell  
15 according to the invention having an electrode in the form of a rotating cylinder:

Figure 1 shows a plot for copper and cadmium deposition at different revolution rates for the rotating cylinder;  
20

Figure 2 shows the variation in depletion rate at different potentials;

Figure 3 is a schematic cross-section of a first embodiment of an  
25 electrical device according to the invention;

Figure 4 is a schematic plan view of a second embodiment of an electrical device according to the invention; and

30 Figure 5 is a schematic plan view of a bipolar plate according to the invention.

Figure 3 shows an electrical device 1 which has a block of electrolyte 2 with asymmetric electrodes 3a,3b at each side of it. The asymmetric electrodes are each provided with electrical connectors 4a,4b. The electrodes are asymmetric electrodes in that one acts as a cathode and the other acts as an anode. The asymmetric electrodes are formed from.

Figure 4 shows an electrical device in the form of a fuel cell 10 having a hydrogen inlet 30 and outlet 35 and an oxygen inlet 40 and outlet 45. The fuel cell 10 has electrodes 20,25 and bipolar plates 15. A bipolar plate 15 is shown in more detail in Figure 5 as having grooves 50 on its surface. The reverse face of the plate 15 also has grooves 50. A variant of the bipolar plate 15 is a separator plate which has grooves 50 on one face only.

The following examples illustrate how to prepare an electrode for use in the invention. The benefits of the invention are also demonstrated. In these examples the materials used are a conventional alkaline phenolic resole (resin A), butyrolactone (an ester), and graphite (conductivity promoter).

#### EXAMPLE 1

This example describes preparation of an alkaline phenol-formaldehyde resin with a formaldehyde to phenol molar ratio of 2.0:1 and a sodium hydroxide to phenol molar ratio of 0.65:1. Phenol (5.0 mol) and sodium hydroxide (0.1 mol) were charged to a reaction vessel and the temperature maintained at 65°C whilst 50% formalin (3.0 mol) was added. The temperature was allowed to be raised to 80°C and maintained at 80°C while a second charge of 50% formalin (5.0 mol) was added slowly over 30 minutes. The mixture was then held at 80°C for 60

minutes before 50% sodium hydroxide solution (3.15 mol) was charged maintaining temperature at 80°C. The resin was condensed at 80°C to a viscosity of 400cP.

5

### EXAMPLE 2

A carbon doped ester-cured resole resin for use as an electrode was prepared by mixing 50g of resin A with 50g of graphite and 10g of butyrolactone in a paper cup. Part of the mixture was poured into a latex  
10 mould and allowed to harden. A gel time was recorded from the mixture left in the cup.

### EXAMPLE 3

15 The cast specimen obtained from Example 2 was allowed to stand at room temperature over 24 hours before conductivity measurements were made using a Como DT3800 Digital Multimeter. The conductivity of the specimen was found to be  $1.12 \times 10^{-1} \text{ S cm}^{-1}$  at 20°C

20

### EXAMPLE 4

This example demonstrates how the resin prepared in Example 2 functions as an electrode material for efficient recovery of metal ions from solution. The resin was made into a cylinder and by the use of silver  
25 epoxy resin was secured to a Rotating Disc Electrode to produce a Rotating Cylinder Electrode, RCE.

This RCE was then used for various experiments, mainly to suggest how efficient the electrode material would be under test conditions. Firstly,  
30 the electrode was used to obtain a current - potential curve for the

deposition of a 1 mmol  $\text{Cu}^{2+}$  and  $\text{Cd}^{2+}$  in 0.5 M  $\text{Na}_2\text{SO}_4$  at pH 2. The results are shown in Figure 1.

5 In this scan, the potential is swept (at  $1 \text{ mVs}^{-1}$ ) and the current is recorded and depends on the process that is occurring, in this case copper and cadmium deposition. The initial curve (in the range of from -150 to -750 mV) is caused by the deposition of copper on the electrodes surface, from -750 to -1050 mV Cadmium has begun to deposit and after -1050 mV hydrogen evolution (as a secondary process) has commenced.

10

From Figure 1, it is clear that the resin according to Example 2 is functioning as an electrode as copper and cadmium is being removed from the mixed solution.

15

#### EXAMPLE 5

In this Example, the electrode of Example 2 was used to collect copper from a solution over a period of 2 hours, and at intervals samples were taken to analyse the amount of copper within the solution. Graph 2 and 3  
20 show results from this experiment.

Firstly from Figure 2, as the potential is increased during the experiment the rate at which the copper is removed from the solution increases, as expected. Also, for the same potential (-0.43 V), if the electrode is  
25 already covered in copper (pre-treated), the rate at which the copper is removed also increases to a similar rate to that when the experiment is ran at -0.60 V. It is shown that either by pre-treating the electrode or increasing the potential during the experiment increases the rate of removal by an extra 10 percent. Over a 2 hour period, the amount of  
30 copper removed is just under 50 %.

These results suggest that the polymer material can be used for metal ion removal from water.

### EXAMPLE 6

5

Carbon doped ester cured phenolic resin electrode material was prepared from carbon graphite grades of varying particle size distribution.

10

Sample A-100g of Graphite grade KL96/97, a 96-97% Carbon flake graphite ground to a d50 of 20-25 microns from Branwell, UK, was mixed with 40 g resin (from example 1), 75 g of water, 0.6 g guar gum thickener and 8g triacetin curing agent. The mixture was mixed in a Kenwood Chef for 2 minutes then poured into 10ml cylindrical moulds and allowed to harden at room temperature. After 15 minutes the specimens were removed from the moulds. Resistivity measurements commenced 1 hour after release from the mould and continued over the next few days.

20

Sample B- As A using coarser grade of graphite grade 2300 d50 36-42 microns

25

Sample C- As A using flake grade graphite KFL96/97, a "small flake" which typically has some 35-40% > 100 microns and 40% < 75 microns, i.e. a mean particle size around 80-90 microns.

Time (hours) after release from mould	Resistance Ohms cm <sup>-1</sup>		
	A	B	C
0	1200	1000	5500
3	180	117	120

6	57	34	62
24	19	23	13
48	7	11	8
72	5	9	7

Table 1 shows that all samples give a high initial resistance and that as curing proceeds the resistance drops rapidly over the next few hours reaching  $< 10 \text{ Ohms cm}^{-1}$ .

5

### EXAMPLE 7

Carbon doped ester cured phenolic resin electrode material was prepared from carbon graphite mixed with other conductive fillers.

Sample D- 40g of graphite grade 2300 from Branwell, UK, was mixed 80g of Copper particles and added to 20g resin (from example 1). 4g triacetin curing agent was added and the mixture mixed in a cup for 2 minutes then poured into 10ml cylindrical mould and allowed to harden at room temperature. After 15 minutes the specimens were removed from the moulds. Resistivity measurements commenced 1 hour after release from the mould and continued over the next few days.

Sample E- As D using Nickel particles 36-42 microns

Time after release from mould	Resistance Ohms $\text{cm}^{-1}$	
	D	E
0	1200	1000
3	70	120
6	61	83

24	40	25
48	21	15
72	14	12

Table 2 shows that all samples give a high initial resistance and that as curing proceeds the resistance drops rapidly over the next few hours reaching  $< 20 \text{ Ohms cm}^{-1}$ .

5

### EXAMPLE 8

Preparation of carbon-doped ester cured phenolic 3-d foam structure

A carbon-doped ester cured phenolic 3d structure was prepared by pre-  
 10 mixing 50 parts phenolic foam resin IDP292 supplied by Borden Chemical  
 UK Ltd with 2 parts DC193 silicone oil supplied by Dow Corning. 100  
 parts KL96/97 graphite powder and 50 parts water were then mixed with  
 the resin until a smooth blend is achieved. Using a high-speed  
 laboratory mixer 10 parts of HCFC141b blowing agent was mixed into the  
 15 resin blend to give a smooth emulsion. 15 parts of butyrolactone curing  
 agent was mixed into the emulsion using the high-speed mixer. After 10  
 seconds mixing the foam mixture was transferred to a plastic mould and  
 immediately placed in an oven at  $50^{\circ}\text{C}$  where the foam was allowed to  
 rise and left to harden overnight. On cutting the foam an open cell,  
 20 fine foam structure of density  $265 \text{ kg m}^{-3}$  was measured. A resistance of  
 $1 \text{ kOhms cm}^{-1}$  was measured after 24 hours.

## CLAIMS

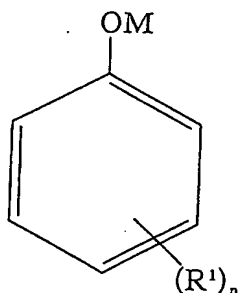
1. An electrode comprising a carbon-doped ester-cured alkaline phenolic resole resin containing conducting alkaline salts.

5

2. An electrode as claimed in claim 1 wherein the ester-cured salt-containing alkaline phenolic resole resin is a reaction product of an ester curing agent with a phenolic resole and a base.

10 3. An electrode as claimed in claim 2 wherein the phenolic resole is a reaction product of a phenol-reactive aldehyde with an alkaline compound of formula

15



(I)

20 wherein R<sup>1</sup> is a straight or branched chain optionally unsaturated alkyl group containing from 1 to 8 carbon atoms (preferably from 1 to 4 carbon atoms, more preferably from 1 to 2 carbon atoms) optionally substituted by a halogen atom (preferably chlorine) or a hydroxy group, a  
25 halogen atom (preferably chlorine), a hydroxy group, and/or a phenyl or benzyl group (optionally substituted by a hydroxy group and/or a straight or branched chain alkyl group containing from 1 to 8 carbon atoms (preferably from 1 to 4 carbon atoms, more preferably from 1 to 2 carbon atoms) optionally substituted by a halogen atom (preferably chlorine) or a hydroxy group);

30 M is a mixture of hydrogen ions and at least one further cation (preferably the at least one further cation is an alkali metal cation



(preferably sodium, lithium or potassium), an alkaline earth metal cation (preferably barium, magnesium or calcium), and/or a  $N(R^2)_4^+$  ion (wherein each  $R^2$  is the same or different and is hydrogen or a straight or branched chain alkyl group containing from 1 to 4 carbon atoms)) wherein  
 5 the molar ratio of hydrogen ions to the at least one further cation is sufficient for the pH to be greater than 7 and is preferably from 2:1 to 1:1; and  
 n is from 0 to 2.

10 4. An electrode as claimed in claim 3 wherein the phenol-reactive aldehyde is a compound of formula



15 wherein R represents hydrogen atom or a straight or branched chain alkyl group having from 1 to 8 (preferably from 1 to 4, more preferably from 1 to 2, most preferably 1) carbon atoms; or a precursor of a compound of formula (II).

20 5. An electrode as claimed in claim 3 or claim 4 wherein the phenol-reactive aldehyde is reacted with the compound of formula (I) in a ratio of from 1:1 to 1:3, preferably from 1:1.2 to 1:3, more preferably from 1:1.5 to 1:3.

25 6. An electrode as claimed in any of claims 2 to 5 wherein the ester curing agent is of formula



30 wherein  $R^3$  represents a hydrogen atom or a straight or branched chain alkyl group containing from 1 to 8 carbon atoms (preferably from 1 to 4,

more preferably from 1 to 2 carbon atoms) optionally substituted by a halogen atom; and

5  $R^4$  represents a straight or branched chain alkyl group containing from 1 to 8 carbon atoms (preferably from 1 to 4, more preferably from 1 to 2 carbon atoms) optionally substituted by one or more hydroxy and/or  $R^3\text{COO-}$  groups, or

10 a phenyl group optionally substituted by a straight or branched chain optionally unsaturated alkyl group containing from 1 to 8 carbon atoms (preferably from 1 to 4 carbon atoms, more preferably from 1 to 2 carbon atoms) optionally substituted by a hydroxy group, a halogen atom (preferably chlorine), a hydroxy group, and/or a phenyl or benzyl group (optionally substituted by a hydroxy group and/or a straight or branched chain alkyl group containing from 1 to 8 carbon atoms (preferably from 1 to 4 carbon atoms, more preferably from 1 to 2 carbon atoms)); or

15  $R^3$  represents a chemical bond to  $R^4$  and  $R^4$  represents a straight or branched chain alkyl group containing from 2 to 10 carbon atoms (preferably from 2 to 4 carbon atoms).

7. An electrode as claimed in any preceding claim wherein the resin  
20 includes a plasticiser to increase flexibility of the resin.

8. An electrode as claimed in any one of the preceding claims wherein the resin to carbon ratio is in the range 0.002-20 : 1.

25 9. An electrode as claimed in claim 8 wherein the carbon is in the form of graphite or an amorphous carbon.

10. An electrode as claimed in claim 9 wherein the carbon is in the form of graphite or an amorphous carbon combined with non-carbon  
30 conductive fillers.

11. An electrode substantially as hereinbefore described and/or as illustrated with reference to Figures 3 and/or 4 of the drawings.
12. A separator plate suitable for use in a fuel cell having one or more  
5 flow field for directing gas flow wherein the plate comprises a carbon-doped ester-cured alkaline phenolic resole resin containing conducting alkaline salts.
13. A separator plate as claimed in claim 12 wherein the resin is as  
10 defined in any one of claims 2 to 10.
14. A separator plate as claimed in claim 12 or claim 13 which has two flow fields.
- 15 15. A separator plate substantially as hereinbefore described and/or as illustrated with reference to Figures 4 and/or 5 of the drawings.
16. An electrical device comprising:  
    (a) a negative electrode;  
20      (b) a positive electrode; and  
    (c) an electrolyte means; and optionally  
    (d) a separator or bipolar plate;  
wherein one or more electrode and/or separator or bipolar plate comprises  
a carbon-doped ester-cured alkaline phenolic resole resin containing  
25 conducting alkaline salts.
17. An electrical device as claimed in claim 16 wherein the resin is as defined in any one of claims 2 to 10.
- 30 18. An electrical device as claimed in claim 16 or claim 17 wherein the separator plate is as defined in any one of claims 12 to 15.

19. An electrical device as claimed in any one of claims 16 to 18 which is a cell, a battery including two or more cells, or a capacitor.
- 5 20. The electrical device as claimed in any one of claims 16 to 19 wherein the electrolyte means is in the form of an electrolyte or it is arranged to receive an electrolyte.
- 10 21. The electrical device as claimed in claim 20 wherein the electrolyte means is in the form of a conduit through which an electrolyte could flow during operation of the cell or in the form of a container into which an electrolyte could be placed at least during operation of the cell.
- 15 22. An electrical device substantially as hereinbefore described and/or as illustrated with reference to Figures 3 to 5 of the drawings.
23. Use of foamed 3-d form of a carbon doped ester-cured alkaline phenolic resole resin containing conducting alkaline salts as an electrode.
- 20 24. Use of a carbon-doped ester-cured alkaline phenolic resole resin containing conducting alkaline salts as an electrode.

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Figure 1

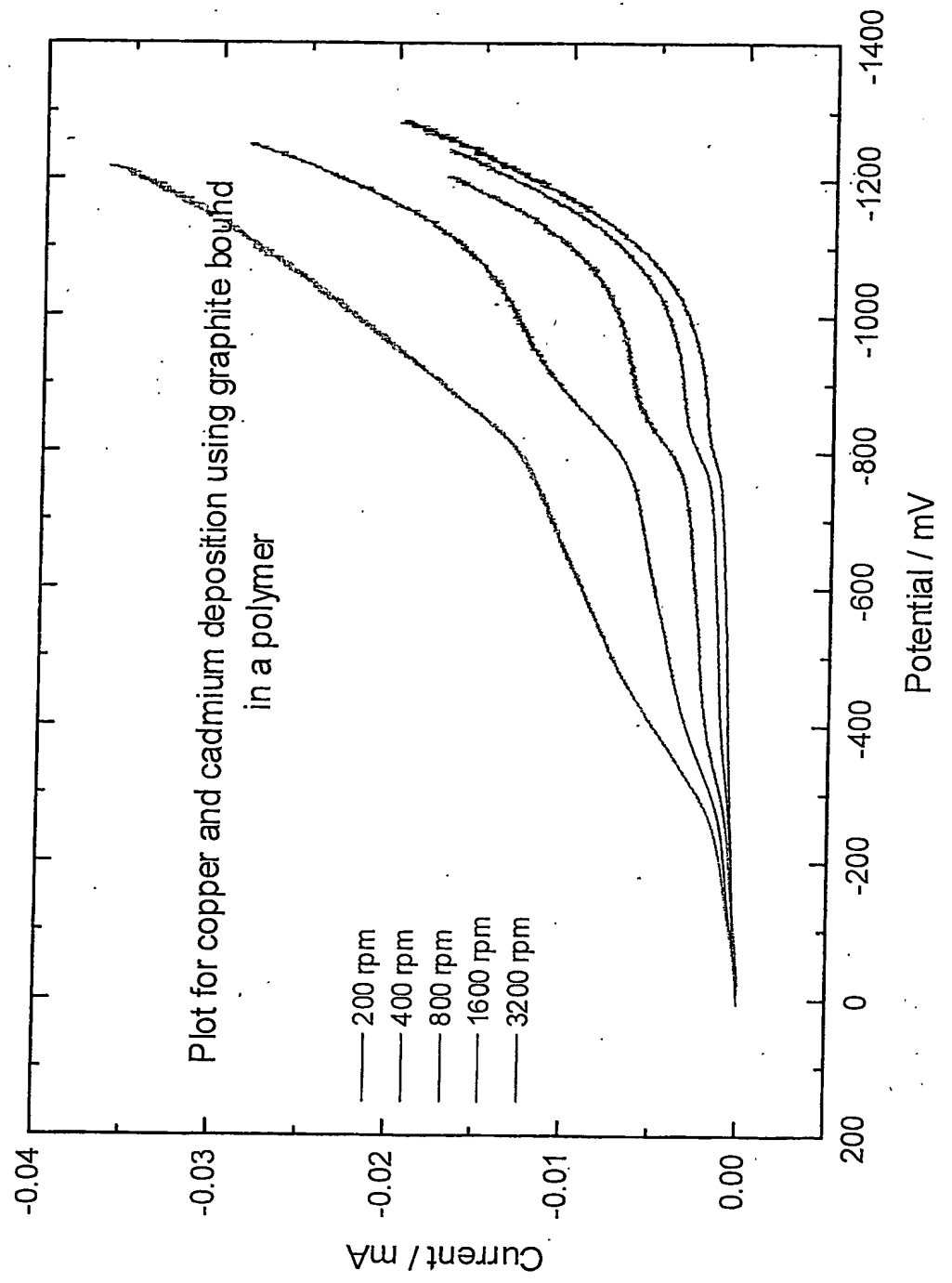
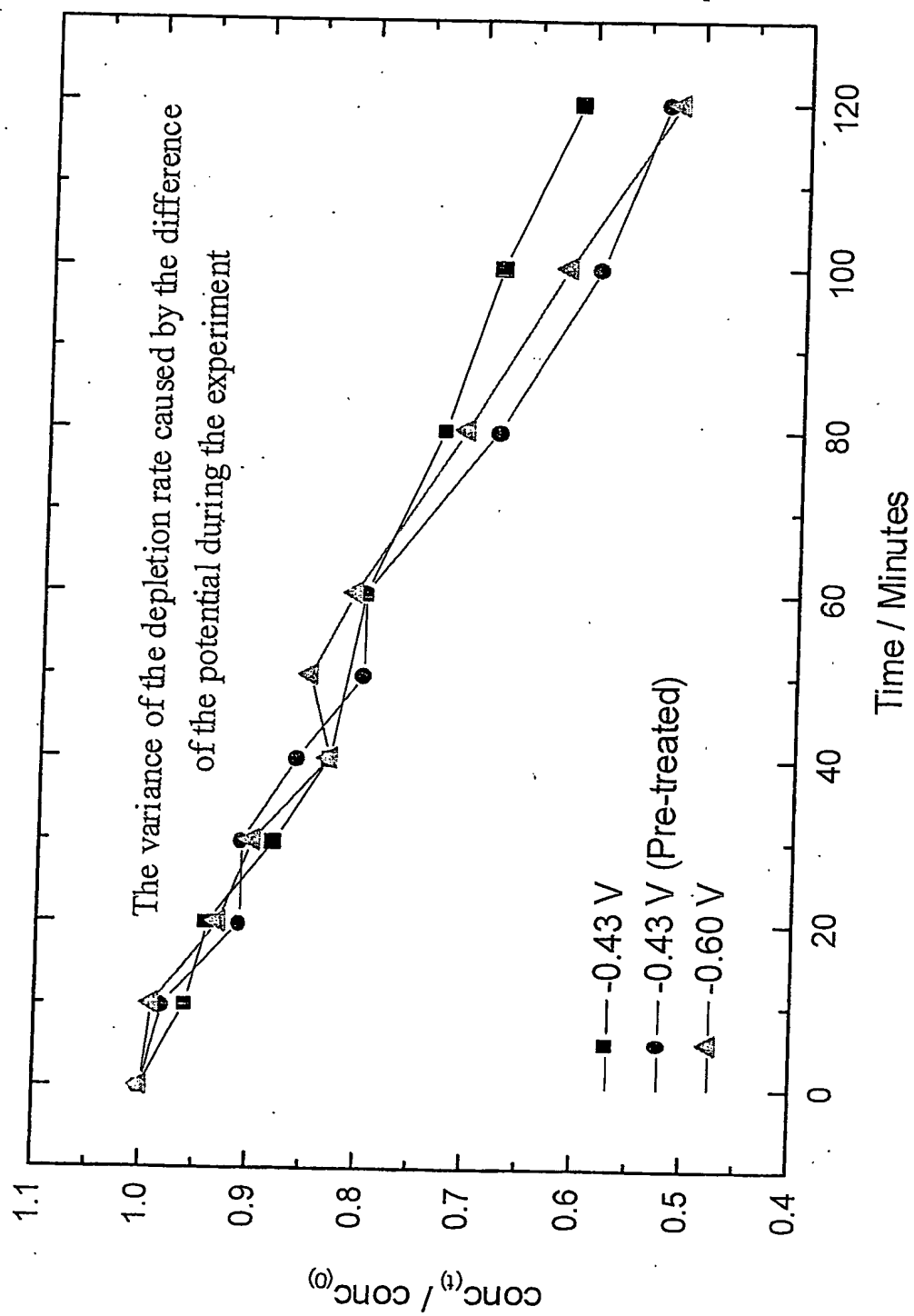
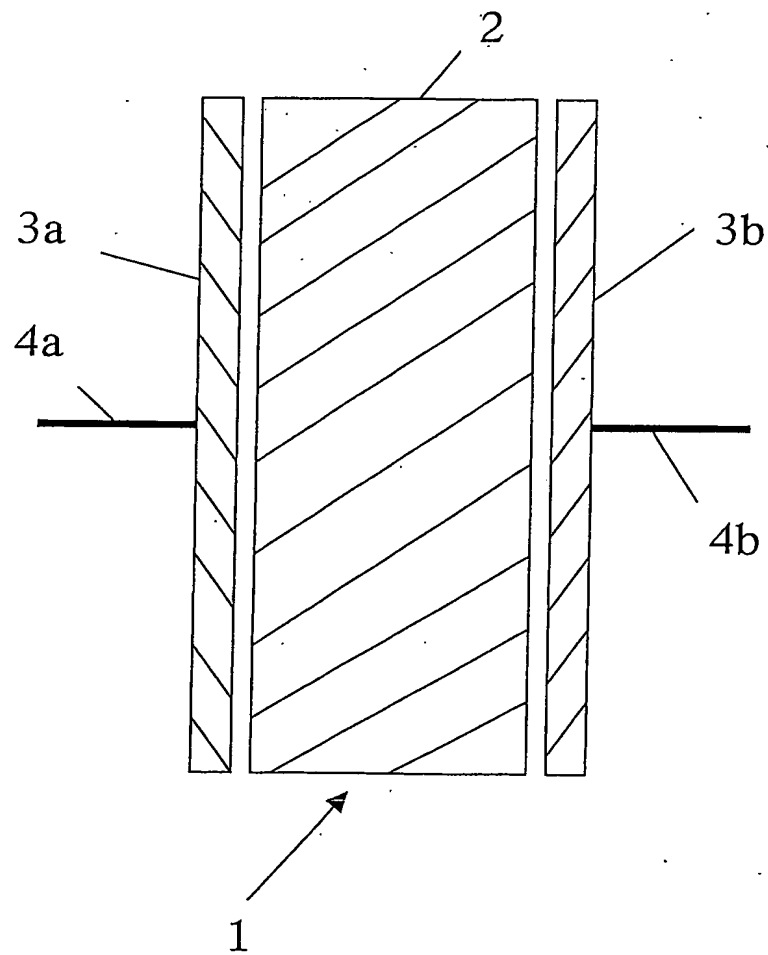


Figure 2



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Figure 3



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Figure 4

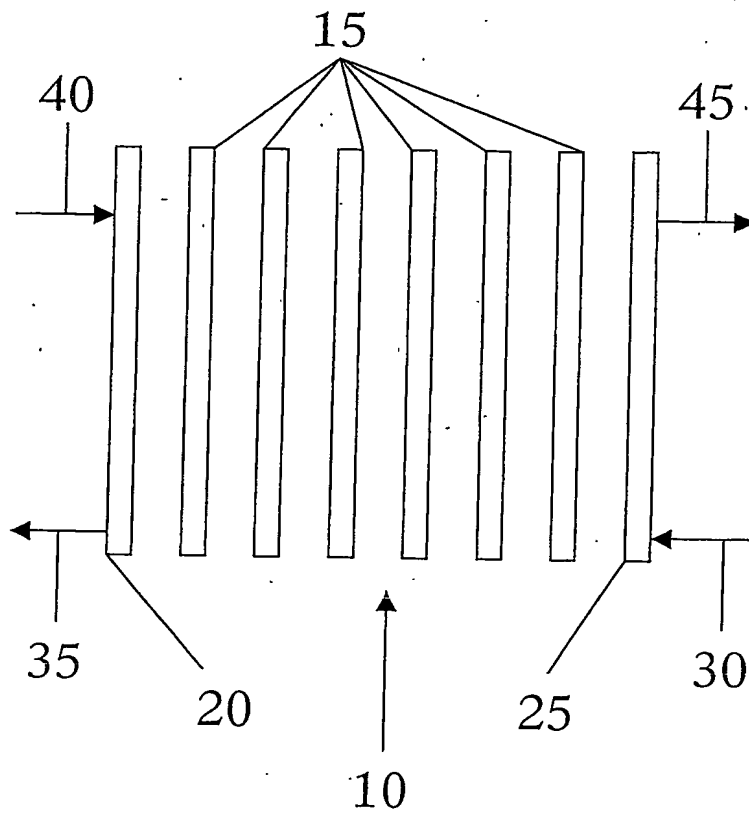
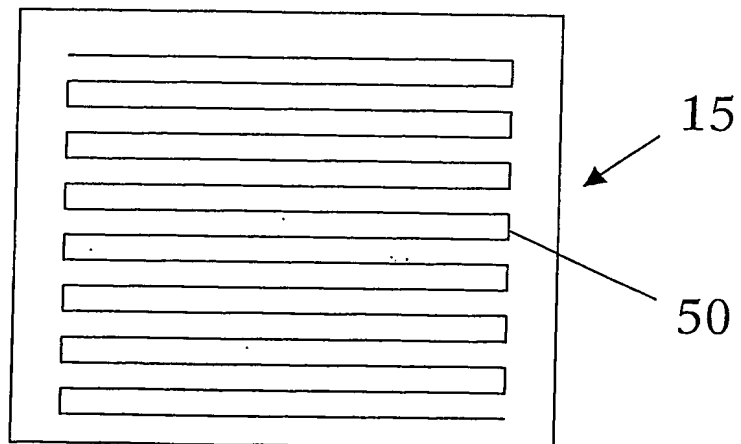


Figure 5





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